EFFECTS OF COOPERATIVE ATOMIC BEHAVIOR ON LASERS

Annual Report

(First year)

by

I.R. Senitzky

September 1978

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CONTENTS

I. Introduction

The purpose of the present discussion is the analysis of the effect of cooperative behavior in those processes of a laser in which atomic coope- ration is usually absent, namely, in pumping and in relaxation. Although lasers have been pumped with laser beams - in particular, in the operation of certain dye lasers - coherent relaxation does not appear to have been considered (in fact, "coherent relaxation" may even sound like a contra- diction in terms). In order to concentrate only on the new aspects of the ideas involved, we will investigate a simple and idealized model.

Consider a collection of identical three-level atoms coupled identically to the field and to the pump. The "field", in this case consists of two resonant modes, the frequencies of which are, respectively, those corresponding to the upper pair of levels and lower pair of levels, *as indicated in Fig. 1.*
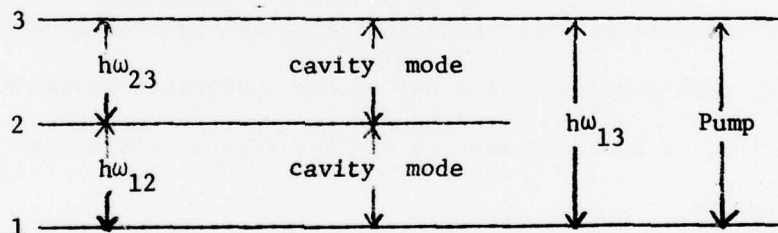

Fig. 1.

The fundamental difference between this system and an ordinary three- level laser is the fact that the transition which would be an incoherent relaxation transition in the ordinary laser can now be a cooperative radiative transition (similar to the "lasing" transition) if such behavior is possible.

We will use the formalism developed previously for cooperative atomic behavior, this formalism being suitable for both quantum mechanical and classical interpretation[1]. The Hamiltonian for the collection of atoms is given by

$$H_o = \sum_{i=1}^{3} h\omega_i a_i^\dagger a_i , \qquad (1.1)$$

where the $a_i^\dagger$'s and a_i's are, for quantum mechanical interpretation, (boson) creation and annihilation operators for atoms in the i'th level, and for classical interpretation, complex harmonic oscillator amplitudes. As previously[1], we use, for convenience, the "reduced" variables A_i, $A_i^\dagger$, defined by

$$a_j(t) = A_j(t)e^{-i\omega_j t} , \quad a_j^\dagger(t) = A_j^\dagger(t)e^{i\omega_j t} . \qquad (1.2)$$

Similarly, the creation and annihilation - or radiation-oscillator - variables for the cavity fields will be denoted by $b_{ij}^\dagger(t)$ and $b_{ij}(t)$, where the pair of subscripts indicates the pair of levels to which the cavity mode couples (the order of the subscripts having no significance); here too, it is convenient to use the reduced variables B_{jk} , $B_{jk}^\dagger$, defined by

$$b_{jk}(t) = B_{jk}(t)e^{-i\omega_{jk}t} , \quad b_{jk}^\dagger(t) = B_{jk}^\dagger e^{i\omega_{jk}t} , \qquad (1.3)$$

where $\omega_{jk} \equiv |\omega_j - \omega_k|$. The part of the Hamiltonian describing the interaction of the atoms with the two cavity modes is given (within the rotating wave approximation) by

$$H_{23}' = h\gamma_{23}(A_2 A_3^\dagger B_{23} + B_{23}^\dagger A_3 A_2^\dagger) , \qquad (1.4)$$

$$H'_{12} = h\gamma_{12}(A_1 A_2^\dagger B_{12} + B_{12}^\dagger A_2 A_1^\dagger) \ . \tag{1.5}$$

The fields of the (lossy) cavity modes may be expressed by[2]

$$B_{12} \simeq B_o^{(12)} - i\gamma_{12} \int_0^t dt' A_1^\dagger(t') A_2(t') e^{-\xi_{12}(t-t')} \ , \tag{1.6a}$$

and

$$B_{23} \simeq B_o^{(23)} - i\gamma_{23} \int_0^t dt' A_2^\dagger(t') A_3(t') a^{-\xi_{23}(t-t')} \ , \tag{1.6b}$$

where $B_o^{(ij)}$ is the cavity field in absence of the molecules, ξ_{ij} is the ij-mode loss constant, and the coupling between field and atoms is assumed to begin at $t = 0$. The pumping may be described by a term in the Hamiltonian

$$H'_{13} = -ih(\mathcal{B} A_1 A_3^\dagger - \mathcal{B}^* A_3 A_1^\dagger) \ , \tag{1.7}$$

where $\mathcal{B}$ is a prescribed function of time. If the pumping is produced by an electromagnetic field, then the field, in appropriate units, is given by

$$\widetilde{\mathcal{B}}(t) = \mathcal{B}(t) e^{-i\omega_{13}t}, \quad \mathcal{B}(t) = \mathcal{B}^*(t) e^{i\omega_{13}t} \ ; \tag{1.8}$$

in other words, $\mathcal{B}$ is also a "reduced" quantity. If the pumping is accomplished by some other method (collisions, for example) then H'_{13} may be regarded as a phenomenological term that represents, approximately, the effect of the pumping.

II. Equations of Motion

Noting that the commutation relations for the atomic variables are $[A_i, A_j^\dagger] = \delta_{ij}$, with all other commutators of the atomic variables vanishing, we obtain, using as the interaction Hamiltonian $H_{12}' + H_{23}' + H_{13}'$,

$$\dot{A}_1 = \mathcal{B}^* A_3 - i\gamma_{12} B_{12}^\dagger A_2 ,$$

$$\dot{A}_2 = -i\gamma_{23} B_{23}^\dagger A_3 - i\gamma_{12} A_1 B_{12} , \qquad (2.1)$$

$$\dot{A}_3 = -\mathcal{B} A_1 - i\gamma_{23} A_2 B_{23} .$$

As will be seen later, it is useful to introduce another set of variables,

$$n_i = A_i^\dagger A_i , \quad S_{ij} = A_i^\dagger A_j \quad \text{for } i \neq j . \qquad (2.2)$$

The equations of motion for these variables follow from those for the A_i's and are given by

$$\dot{n}_1 = \mathcal{B}^* S_{13} - i\gamma_{12} B_{12}^\dagger S_{12} + \text{H.C.} ,$$

$$\dot{n}_2 = -i\gamma_{23} B_{23}^\dagger S_{23} - i\gamma_{12} S_{12}^\dagger B_{12} + \text{H.C.},$$

$$\dot{n}_3 = -\mathcal{B} S_{13}^\dagger - i\gamma_{23} S_{23}^\dagger B_{23} + \text{H.C.} ,$$

$$\dot{S}_{12} = \mathcal{B} S_{23}^\dagger + i\gamma_{12}(n_2 - n_1) B_{12} - i\gamma_{23} B_{23}^\dagger S_{13} , \qquad (2.3)$$

$$\dot{S}_{23} = -\mathcal{B} S_{12}^\dagger + i\gamma_{23}(n_3 - n_2) B_{23} + i\gamma_{12} B_{12}^\dagger S_{13} ,$$

$$\dot{S}_{13} = \mathcal{B} (n_3 - n_1) + i\gamma_{12} B_{12} S_{23} - i\gamma_{23} S_{12} B_{23} .$$

The n_i and S_{ij} variables have more direct physical meaning than the A_i's , since they refer to level populations and dipole moments. For the case of maximum atomic cooperation, the equations for the two sets of variables are essentially equivalent (and valid both classically and quantum mechanically). Equations (2.3) are, however, more suitable for applying the formalism to the case of incoherent pumping, for modification of the formalism to the

case of less than maximum cooperation, and for the introduction of phenomenological relaxation constants (which may, in turn, affect cooperation).

Since our interest lies in macroscopic phenomena, our first step in proceeding from Eqs. (2.3), is the consideration of the dynamical variables to be classical variables. In this format, we consider the case of incoherent pumping. The function $\mathcal{B}(t)$ is taken to be a gaussian random process. For simplicity, we consider maximum incoherence ("white noise"), defined by

$$\left\langle \mathcal{B}(t) \right\rangle = 0 \ , \quad \left\langle \mathcal{B}^*(t_1)\mathcal{B}(t_2) \right\rangle = 2p\delta(t_1-t_2) \ , \tag{2.4}$$

$$\left\langle \mathcal{B}(t_1)\mathcal{B}(t_2) \right\rangle = \left\langle \mathcal{B}^*(t_1)\mathcal{B}^*(t_2) \right\rangle = 0 \ , \tag{2.5}$$

with the angular bracket indicating an average over members of an ensemble associated with the statistical description of the random process. Let us calculate the average of all terms in Eqs. (2.3) that contains the factor $\mathcal{B}$. One obtains, by means of Eqs. (2.1),

$$
\begin{aligned}
\left\langle \mathcal{B} \, s_{23}^\dagger \right\rangle &= \left\langle \mathcal{B} \, A^*_3 A_2 \right\rangle \\
&= \left\langle \mathcal{B}(t)\{A_3^*(0) - \int_0^t dt_1 [\mathcal{B}^*(t_1)A_1^*(t_1) \right. \\
&\qquad\qquad \left. + i\gamma_{23}A_2^*(t_1)B_{23}^*(t_1)]\}A_2(t) \right. \\
&= -\int_0^t dt_1 \left\langle \mathcal{B}(t)\mathcal{B}^*(t_1) \right\rangle A_1^*(t_1)A_2(t) \ , \\
&= -ps_{12} \ , \tag{2.6}
\end{aligned}
$$

where terms of higher order than the second in either the coupling constant γ_{ij}, or $\mathcal{B}$ (in which a coupling constant is already included), or both have been dropped for purposes of substitution back into the equations of motion. Similarly, one obtains

$$\langle \mathcal{B} s_{12}^* \rangle \simeq p S_{23} \,, \tag{2.7}$$

$$\langle \mathcal{B} (n_3 - n_1) \rangle \simeq - 2 p S_{13} \,. \tag{2.8}$$

Using the above approximation <u>and</u> a random phase approximation, one obtains, also,

$$\langle \mathcal{B} s_{13} \rangle \simeq p(n_3 - n_1) \,. \tag{2.9}$$

We approximate the equations of motion (2.3) by replacing all terms in these equations that contain the factor $\mathcal{B}$ by their averages. The result is

$$\dot{n}_1 = 2p(n_3 - n_1) - i\gamma_{12}B_{12}^* S_{12} + i\gamma_{12}B_{12}S_{12}^* \,,$$

$$\dot{n}_2 = - i\gamma_{23}B_{23}^* S_{23} + i\gamma_{23}B_{23}S_{23}^* - i\gamma_{12}B_{12}S_{12}^* + i\gamma_{12}B_{12}^* S_{12} \,,$$

$$\dot{n}_3 = - 2p(n_3 - n_1) - i\gamma_{23}B_{23}S_{23}^* + i\gamma_{23}B_{23}^* S_{23} \,,$$

$$\dot{S}_{12} = - p S_{12} + i\gamma_{12}(n_2 - n_1)B_{12} - i\gamma_{23}B_{23}^* S_{13} \,, \tag{2.10}$$

$$\dot{S}_{23} = - p S_{23} + i\gamma_{23}(n_3 - n_2)B_{23} + i\gamma_{12}B_{12}^* S_{13} \,,$$

$$\dot{S}_{13} = - 2p S_{13} + i\gamma_{12}B_{12}S_{23} - i\gamma_{23}B_{23}S_{12} \,.$$

One notes that incoherent pumping, within the approximation framework used, has the effect of several rate processes, transferring atoms from level 1 to 3 and vice versa at the same rate per atom, and attenuating the dipole moments at certain rates. The result appears physically reasonable, exhibiting the damping effect of an incoherent perturbation on a macroscopic oscillation (on the average).

In the case of coherent pumping exactly on the ω_{13} resonance, $\mathcal{B}$ is constant. We have, therefore, two sets of classical equations: Eqs. (2.3) with $\mathcal{B}$ constant for coherent pumping, and Eqs. (2.10) for incoherent pumping.

We turn next to the case of less than maximum cooperation. The formalism may be generalized to this case by considering the S_{ij}'s and n_i's to be independent variables, subject only to the relationship

$$|S_{ij}|^2 \le n_i n_j , \qquad (2.11)$$

with the equality sign corresponding to maximum cooperation. [It is interesting to note that the approximations used to obtain Eqs. (2.10) for incoherent pumping have accomplished this already, in a special manner.] One may now introduce phenomenological relaxation constants for both the energy and the dipole moment. Eqs. (2.3) then become

$$\dot{n}_1 = \mathcal{B}^* S_{13} - i\gamma_{12}B_{12}^* S_{12} + c.c. + \beta_{31}n_3 + \beta_{21}n_2$$

$$\dot{n}_2 = - i\gamma_{23}B_{23}^* S_{23} - i\gamma_{12}B_{12}S_{12}^* + c.c. + \beta_{32}n_3 - \beta_{21}n_2$$

$$\dot{n}_3 = - \mathcal{B}S_{13}^* - i\gamma_{23}B_{23}S_{23}^* + c.c. - \beta_{32}n_3 - \beta_{31}n_3$$

$$\dot{S}_{12} = \mathcal{B}S_{23}^* + i\gamma_{12}(n_2-n_1)B_{12} - i\gamma_{23}B_{23}^* S_{13} - \alpha_{12}S_{12} \qquad (2.12)$$

$$\dot{S}_{23} = - \mathcal{B}S_{12}^* + i\gamma_{23}(n_3-n_2)B_{23} + i\gamma_{12}B_{12}^* S_{13} - \alpha_{23}S_{23}$$

$$\dot{S}_{13} = \mathcal{B}(n_3-n_1) + i\gamma_{12}B_{12}S_{23} - i\gamma_{23}S_{12}B_{23} - \alpha_{13}S_{13}$$

These equations may be regarded as a generalization of the Bloch equations (which are suitable for a collection of two-level atoms) to a collection of three-level atoms, the β's being "longitudinal" relaxation constants and the α's "transverse" relaxation constants. The same method can be used, of course, to obtain equations for a collection of atoms with more than three levels.

For the case of incoherent pumping, Eqs. (2.10) already include transverse relaxation constants. Although these have special values determined only by the pumping strength, we will, for the sake of simplicity, neglect other transverse relaxation processes (assuming them to be small compared to the effect of the pumping) and insert only longitudinal relaxation constants. Here, too, however, we will ignore β_{31} compared to p, so that the equations of motion for incoherent pumping become

$$\dot{n}_1 = 2p(n_3-n_1) - i\gamma_{12}B_{12}^*S_{12} + i\gamma_{12}B_{12}S_{12}^* + \beta_{21}n_2 ,$$

$$\dot{n}_2 = - i\gamma_{23}B_{23}^*S_{23} + i\gamma_{23}B_{23}S_{23}^* - i\gamma_{12}B_{12}S_{12}^*$$

$$+ i\gamma_{12}B_{12}^*S_{12} - \beta_{21}n_2 + \beta_{32}n_3 ,$$

$$\dot{n}_3 = - 2p(n_3-n_1) - i\gamma_{23}B_{23}S_{23}^* + i\gamma_{23}B_{23}^*S_{23} - \beta_{32}n_3 ,$$

$$\dot{S}_{12} = - pS_{12} + i\gamma_{12}(n_2-n_1)B_{12} - i\gamma_{23}B_{23}^*S_{13} , \qquad (2.13)$$

$$\dot{S}_{23} = - pS_{23} + i\gamma_{23}(n_3-n_2)B_{23} + i\gamma_{12}B_{12}^*S_{13} ,$$

$$\dot{S}_{13} = - 2pS_{13} + i\gamma_{12}B_{12}S_{23} - i\gamma_{23}B_{23}S_{12} .$$

In addition to the equations of motion (2.12) or (2.13) we have the equation that is consistent with either set, namely, the statement of the conservation of the number of atoms,

$$n_1 + n_2 + n_3 = N , \qquad (2.14)$$

N being the total number of atoms under consideration. (Note the $\sum \dot{n}_i = 0$ in all of the above equations.)

III. Steady-State Equations

In the present analysis, our interest will be confined to the steady-state situation only. In this situation all the (reduced) variables become constants. The expressions for the field, Eqs. (1.6), may be integrated immediately to yield

$$B_{jk} = B_o^{(jk)} - i(\gamma_{jk}/\xi_{jk})S_{jk} . \tag{3.1}$$

If we are interested in the case where the cavity fields are due to the atomic dipole moments only, in other words, if fields due to external sources are absent, then $B_o^{(jk)} = 0$. Setting the time derivatives in the equations of motion equal to zero, substituting for B_{jk} from Eq. (3.1), and using the notation

$$z_{ij} \equiv \gamma_{i\gamma}^2/\xi_{ij} , \tag{3.2}$$

the equations of motion become, for coherent pumping

$$\mathcal{B}^* s_{13} + \mathcal{B} s_{13}^* + 2z_{12}|S_{12}|^2 + \beta_{21}n_2 = 0$$

$$2z_{23}|S_{23}|^2 - 2z_{12}|S_{12}|^2 + \beta_{32}n_3 - \beta_{21}n_2 = 0 ,$$

$$\mathcal{B} s_{13}^* + \mathcal{B}^* s_{13} + 2z_{23}|S_{23}|^2 + \beta_{32}n_3 = 0 ,$$

$$(\mathcal{B} + z_{23}s_{13})S_{23}^* + z_{12}(n_2-n_1)S_{12} - \alpha_{12}S_{12} = 0 , \tag{3.3}$$

$$- (\mathcal{B} + z_{12}s_{13})S_{12}^* + z_{23}(n_3-n_2)S_{23} - \alpha_{23}S_{23} = 0 ,$$

$$\mathcal{B}(n_3-n_1) + (z_{12} - z_{23})S_{12}S_{23} - \alpha_{13}S_{13} = 0 ,$$

and, for incoherent pumping

$$2p(n_3-n_1) + 2z_{12}|S_{12}|^2 + \beta_{21}n_2 = 0 \ ,$$

$$2z_{23}|S_{23}|^2 - 2z_{12}|S_{12}|^2 + \beta_{32}n_3 - \beta_{21}n_2 = 0 \ ,$$

$$2p(n_3-n_1) + 2z_{23}|S_{23}|^2 + \beta_{32}n_3 = 0 \ ,$$

$$pS_{12} - z_{12}(n_2-n_1)S_{12} - z_{23}S_{23}^*S_{13} = 0 \ , \tag{3.4}$$

$$pS_{23} - z_{23}(n_3-n_2)S_{23} + z_{12}S_{12}^*S_{13} = 0 \ ,$$

$$2pS_{13} - (z_{12} - z_{23})S_{12}S_{23} = 0 \ .$$

Before seeking solutions of these equations, it is worthwhile to make another simplification, which is not qualitatively significant and fits our original intention of treating the transition between the upper pair of levels and that between the lower pair of levels more or less similarly. Since the quantity z_{ij} may be regarded as a measure of the effect of the cavity on the atoms – and vice versa – with respect to the ij transition, we set

$$z_{12} = z_{23} \equiv z \ . \tag{3.5}$$

For notational simplicity and physical insight, we introduce the quantity

$$P_{ij} = 2z_{ij}|S_{ij}|^2 = 2\xi_{ij}|B_{ij}|^2 \tag{3.6}$$

The rate at which field energy decays in the (free) cavity per unit energy is $2\xi_{ij}$, so that P_{ij} is the rate at which photons are absorbed from the atoms by the cavity. (The analysis is classical, and the photon is to be considered here merely as a unit of energy.) We can simplify Eqs. (3.3) and (3.4) further by noting that only two of the first three equations in each set are independent, so that one of the first three equations, say, the second, may be dropped. On the other hand, each set of

equations must be supplemented by Eq. (2.14). Taking note of all the above comments and simplifications, we obtain the following sets of equations:

For incoherent pumping, we have

$$2p(n_3-n_1) + P_{12} + \beta_{21}n_2 = 0 \ ,$$

$$2p(n_3-n_1) + P_{23} + \beta_{32}n_3 = 0 \ ,$$

$$n_1 + n_2 + n_3 = N \ ,$$

$$S_{12}[p - z(n_2-n_1)] = 0 \ ,$$

$$S_{23}[p - z(n_3-n_2)] = 0 \ ; \tag{3.7}$$

for coherent pumping, we have

$$\mathcal{B}^*S_{13} + \mathcal{B}S_{13}^* + P_{12} + \beta_{21}n_2 = 0$$

$$\mathcal{B}^*S_{13} + \mathcal{B}S_{13}^* + P_{23} + \beta_{32}n_3 = 0$$

$$n_1 + n_2 + n_3 = 0$$

$$(\mathcal{B} + zS_{13})S_{23}^* + [z(n_2-n_1) - \alpha_{12}]S_{12} = 0$$

$$(\mathcal{B} + zS_{13})S_{12}^* - [z(n_3-n_2) - \alpha_{23}]S_{23} = 0$$

$$(n_3-n_1) - \alpha_{13}S_{13} = 0 \ . \tag{3.8}$$

Note that for incoherent pumping the assumption of Eq. (3.5) accounts for the vanishing of S_{13} .

Our main interest, of course, lies in the cavity fields B_{jk} - which are determined by the dipole moments S_{jk} through Eq. (3.1) - or, alternatively, in the cavity powers P_{jk} - which are determined by the dipole moments S_{jk} through Eq. (3.6). The level populations n_i may be regarded as auxiliary variables which are useful for determining

the fields. Both sets of equations are nonlinear, and there are more unknowns than equations, since the S_{ij}'s are complex. One cannot therefore expect unique solutions. We investigate solutions of the two sets of equations separately, looking at the simpler set, that for incoherent pumping, first.

IV. Steady-State Solutions. Incoherent Pumping

A trivial solution of Eqs. (3.7) exists for $S_{12} = S_{23} = 0$. In this case $P_{12} = P_{23} = 0$, and we have three linear inhomogeneous equations for n_1, n_2, n_3 with the solution

$$n_1 = N\beta_{21}(2p + \beta_{32})/D ,$$

$$n_2 = 2Np\beta_{32}/D ,$$ (4.1)

$$n_3 = 2Np\beta_{21}/D ,$$

where

$$D \equiv 2p(2\beta_{21} + \beta_{32}) + \beta_{21}\beta_{32} .$$

This is the steady state population one would obtain in the absence of cavity-coupling for all values of p. In the present case, one can show that this solution becomes unstable for certain value of the parameters. However, we will not discuss the stability of solutions in the present analysis.

We look next for a solution in which either S_{12} or S_{23} does not vanish. For $S_{23} = 0$, Eqs. (3.7) become four linear equations for four unknowns n_1, n_2, n_3, P_{12}, with the solution

$$n_1 = (N-p/z)(2p + \beta_{32})/D ,$$

$$n_2 = n_1 + p/z = [N(2p + \beta_{32}) + (p/z)(4p + \beta_{32})]/D ,$$

$$n_3 = 2p(N - p/z)/D ,$$

$$P_{12} = D^{-1}\{N[2p(\beta_{32} - \beta_{21}) - \beta_{21}\beta_{32}] - (2p^2/z)(2\beta_{21} + \beta_{32})$$
$$- (p/z)\beta_{21}\beta_{32}\}$$

(4.2)

where

$$D \equiv 2(3p + \beta_{32}) .$$

Since all of these four quantities must be positive, it is clear that a solution exists only for certain ranges of the parameters. Thus, we must have $N > p/z$ and $\beta_{32} > \beta_{21}$, and these inequalities must be of sufficient magnitude so that $P_{12} > 0$. For $N \gg p/z$ and $p \sim \beta_{32} \gg \beta_{21}$, one obtains, approximately,

$$P_{12} \approx \frac{Np\beta_{32}}{3p+\beta_{32}} \sim \frac{1}{4} N .$$

(4.3)

Since for $p = 0$, the formal expression for P_{12} is negative, there exists a threshold value for the pumping (given by the lower root of P_{12} as a quadratic in p) and also an upper limit (the upper root) above which a steady state solution does not exist.

A solution for which P_{23} rather than P_{12} does not vanish, obtained by setting $S_{12} = 0$, is given by

$$n_1 = [N(2p + \beta_{21}) + (p/z)(2p - \beta_{21})]/D ,$$

$$n_2 = [2pN - (2p/z)(2p + \beta_{21})]/D ,$$

$$n_3 = n_2 + p/z = [2pN + (p/z)(2p - \beta_{21})]/D ,$$

$$P_{23} = [2pN(\beta_{21} - \beta_{32}) - (2p^2/z)(2\beta_{21} + \beta_{32}) - (p/z)\beta_{21}\beta_{32}]/D ,$$

(4.4)

where

$$D \equiv 6p + \beta_{21} \ .$$

In order for a nonvanishing field to exist (in the ω_{23} cavity), we must have $\beta_{21} > \beta_{32}$, $N > p/z$, and these inequalities must be of sufficient magnitude so that $P_{23} > 0$. For $N \gg p/z$ and $p \sim \beta_{21} \gg \beta_{32}$, one obtains approximately

$$P_{23} \simeq \frac{2pN\beta_{21}}{6p+\beta_{21}} \sim \frac{2}{7} N \ . \tag{4.5}$$

The above two cases of oscillation in only one of the cavities, whether that coupled to the upper pair of levels or that coupled to the lower pair of levels,are essentially the cases of the usual three-level laser. The longitudinal (incoherent) relaxation in the non-oscillating transition is essential for operation, of course.

We ask now: does a solution exist in which the field oscillates in both cavities? It is easy to see from Eqs. (3.7) that such a solution does not exist. If $S_{12} \neq 0$ and $S_{23} \neq 0$, we must have

$$p = z(n_2 - n_1) = z(n_3 - n_2) \ , \tag{4.6}$$

which, since $p > 0$, requires

$$n_3 > n_2 > n_1 \ . \tag{4.7}$$

However, from the first or second of equations (3.7) we must have $n_3 < n_1$, since the second and third terms in these equations are positive. Thus, the simultaneous existence of an oscillating field in both cavities, for the case of incoherent pumping, is impossible. The coupling of a resonant cavity to the "nonlasing" transition produces no effect in an incoherently pumped three-level laser.

Before we leave the case of incoherent pumping, it might be worthwhile to point out that the first two of equations (3.7) have a very simple physical interpretation. They are equations for the rates at which atoms leave and arrive at levels 1 and 3. Consider the first equation for example. The rate at which the incoherent pumping changes the population of level 1 by transferring atoms to level 3 is $-2pn_1$ and the rate of the reverse process is $2pn_3$. The rate at which atoms arrive from level 2 to level 1 by relaxation is $\beta_{21}n_2$, and P_{12} — being the number of photons absorbed by the cavity — is the rate at which atoms arrive from level 2 to level 1 by coherent radiation. (Note that β_{31} was neglected compared to p .)

V. Steady-State Solutions. Coherent Pumping

We consider now the case of coherent pumping. Equations (3.8) for coherent pumping are more complicated than Eqs. (3.7) for incoherent pumping, since the complex variables S_{ij} , and not merely their absolute values [as in the case of Eqs. (3.7)], enter in an essential way in the solutions. In other words, the phase of all the oscillating quantities (dipole moments and fields) becomes significant here. This is hardly surprising since the coherent pumping, described by β , contains phase information itself. Since β is the only prescribed complex quantity, or parameter, we can, without loss of generality, take it to the real and positive. In the present analysis we will consider Eqs. (3.8) not in their full generality, but rather in a simplified form which, nevertheless, illuminates the ideas under investigation. This simplification consists in the neglect of all (incoherent) relaxation processes. Although, in the

case of incoherent pumping, the existence of one of the relaxation processes is an essential requirement for the operation of a three-level laser, it will be seen that, in the present instance, interesting and nontrivial results are obtained in the absence of such processes.

If we drop the relaxation constant and take $\mathcal{B}$ to be real and positive, Eqs. (3.8) become

$$\mathcal{B}(S_{13} + S_{13}^*) + P_{12} = 0 ,$$

$$P_{23} = P_{12} ,$$

$$n_1 + n_2 + n_3 = N ,$$

$$(\mathcal{B} + zS_{13})S_{23}^* + z(n_2 - n_1)S_{12} = 0 ,$$

$$(\mathcal{B} + zS_{13})S_{12}^* - z(n_3 - n_2)S_{23} = 0 ,$$

$$n_1 = n_3 .$$

$$(5.1)$$

Introducing the notation

$$n = n_1 = n_3$$

$$S_{jk} = |S_{jk}|e^{-i\theta_{jk}} ,$$

$$S = |S_{12}| = |S_{23}| ,$$

$$\theta = \theta_{12} + \theta_{23} ,$$

$$(5.2)$$

we can reduce Eqs. (5.1) to

$$zS^2 = -\mathcal{B} \ \text{Re}\{S_{13}\} \tag{5.3a}$$

$$S[(\mathcal{B} + zS_{13}) + z(N - 3n)e^{-i\theta}] = 0 . \tag{5.3b}$$

Equations (5.3) are the only requirements on the variables S, n, S_{13} and θ, except for the restrictions [from Eqs. (2.11)]

$$S^2 \le n(N - 2n) \ , \qquad |S_{13}|^2 \le n^2 \ . \tag{5.4}$$

It is clear that the solution for these variables is not unique, and a number of steady-state conditions are possible for a given β, z, and N , some of which we proceed to examine.

A trivial solution of Eqs. (5.3) is given by

$$S = 0 \ , \qquad Re\{S_{13}\} = 0 \ . \tag{5.5}$$

From Eqs. (2.3), one sees that the power absorbed by the atoms from the pumping field is given by

$$-\beta (S_{13}^* + S_{13}) h\omega_3 + \beta (S_{13}^* + S_{13}) h\omega_1 ,$$
$$= -\beta (S_{13}^* + S_{13}) h\omega_{13} = -2\beta h\omega_{13} Re\{S_{13}\} \ . \tag{5.6}$$

Thus, for $Re\{S_{13}\} = 0$, no power is absorbed from the pump, and naturally, the cavity fields are zero. In this case, S_{13} is a pure imaginary quantity: in other words, although S_{13} need not vanish, it oscillates $\frac{1}{2}\pi$ radians out of phase with the pumping field and no power is absorbed (when averaged over a cycle, the averaging being implicit in the rotating wave approximation).

Equation (5.3a) has a very simple physical meaning. Since $P_{12} = P_{13} = 2zS^2$, this equation states that the number of photons (at the respective frequencies) absorbed by each of the cavities is equal to the number of photons absorbed by the atoms from the pump (at the pumping frequency); in other words, it is the statement of the conservation of energy.

A mathematically simple — but physically significant — solution of Eqs. (5.3) is given by

$$n = \frac{1}{3} N , \quad S_{13} = -\beta/z , \quad S = \beta/z . \tag{5.7}$$

What is most interesting about this solution is the fact that θ is arbitrary. Here we have a situation in which both cavities oscillate with arbitrary phases, that is, the phases of the cavity oscillations are independent of the pump phase. This behavior is qualitatively different from that of a parametric oscillator (or Raman laser), in which the relationship between the signal and idler phase is determined by the phase of the pumping oscillator. One may look at this difference in behavior as an illustration of the difference between a virtual level and a real level, since in a parametric amplifier, as illustrated in Fig. 2,
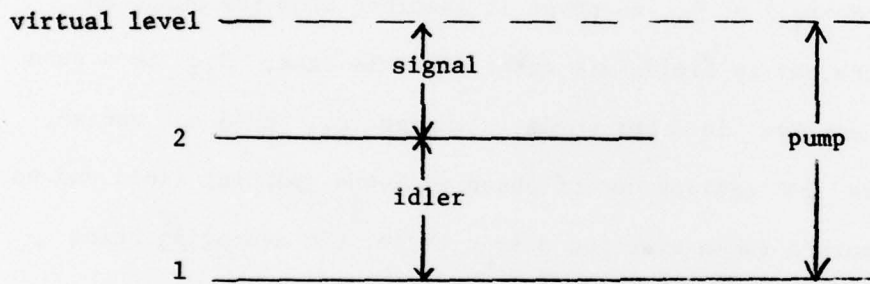

Fig. 2

the pump is often regarded as creating a (third) virtual level. The power dissipated in the cavities depends of course on the strength of the pump β, but one should note that inequalities (5.4) lead to

$$\beta \leq \frac{1}{3} Nz \tag{5.8}$$

the equality sign indicating maximum cooperation. For β greater than that permitted by (5.8), a solution of the form given by Eqs. (5.7) cannot exist. The maximum steady-state power that can be dissipated in each cavity for $n = \frac{1}{3} N$ is

$$P_{max} = \frac{2}{9} zN^2 .$$ (5.9)

In general, Eq. (5.3b) stands for the two real equations

$$|S_{13}| \sin\theta_{13} + (N - 3n)\sin\theta = 0$$ (5.10a)

$$|S_{13}| \cos\theta_{13} + (N - 3n)\cos\theta = -\mathcal{B}/z ,$$ (5.10b)

with $|S_{13}| \leq n$ and $n < \frac{1}{2} N$. For $n \neq \frac{1}{3} N$, there does exist a relationship between θ and θ_{13} , which becomes more critical as $|N - 3n|$ increases from zero. It is of interest to look at the solution that corresponds to the maximum power that may be absorbed by the atoms under steady-state conditions. This will obviously be the solution corresponding to maximum cooperation <u>and</u> maximum S . From Eq. (5.4) , we see that such maximization corresponds to $n = \frac{1}{4} N$, for which

$$P_{max} = \frac{1}{4} zN^2 .$$ (5.11)

Since $|S_{13}| = n$, Eq. (5.10a) yields $\theta = -\theta_{13}$, so that Eq. (5.10b) results in

$$\frac{1}{2} N\cos\theta = -\mathcal{B}/z .$$ (5.12)

On the other hand, Eq. (5.3a) requires

$$zN = -2\mathcal{B}\cos\theta .$$ (5.13)

These two equations can be satisfied only for $\mathcal{B} = \frac{1}{2} zN$, and the solution is

$$\cos\theta = -1 , \quad \text{or} \quad \theta = \pi$$ (5.14)

In this case, the phase relationship between the oscillations at the various frequencies is similar to that for a parametric oscillator, and the value of θ_{13} is that for the greatest absorption of power from the pump (for a given $|S_{13}|$) .

VI. Conclusions

The coupling of resonant cavity modes to both transitions of a
coherently pumped collection of three-level systems leads to behavior
that is different under certain conditions from both that of a laser and
a parametric oscillator. In particular, there can exist oscillations in
both modes that are independent in phase. Such behavior would open
the possibility of obtaining oscillation in an extended medium at both transition
transition frequencies without the need of phase matching, a need that
exists in the case of parametric oscillation (or a Raman laser). The
model studied in the present discussion was simplified by dropping certain
terms in the equations of motion. Further study of a more realistic model,
that is, a study of the solution of the full equations of motion, including
the stability of the steady-state solutions, and perhaps, time dependent
behavior, is necessary for a clearer picture of the potentialities
of coherent pumping and relaxation.

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2. I.R. Senitzky, Phys. Rev. 155, 1387 (1967).